



## DECLARATION

### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Inventors: Kunio Nishimura, Akinori Sudoh, Chiaki Sotowa

Filing Date: April 9, 2001

Application No.: 09/807,075

Group Art Unit: 1754

Examiner: Peter J. Lish

Title: CARBON MATERIAL FOR BATTERY AND BATTERY CONTAINING  
THE CARBON MATERIAL

### DECLARATION PURSUANT TO 37 C.F.R. 1.132

Commissioner for Patents

Washington, D.C. 20231

Sir:

I, Akinori Sudoh, residing in Kanagawa-ken, Japan, declare and state that

1. I graduated from Tokyo University of Agriculture and Technology, Graduate School of Technology, Master's Program for Technology, in March 1989. Since June 2003, I have been employed by SHOWA DENKO K.K., and have been engaged in research in the Fine Carbon Division.

2. I am one of the inventors of the invention as claimed in the above-referenced application, and accordingly, I am familiar with the specification and claims which comprise that application.

3. I am aware of the Final Office Action of August 9, 2004, issued on the

above-referenced application, in which Claims 1-9 and 30-33 were rejected under 35 U.S.C. 103(a) as being unpatentable over Hase et al. (US 5,910,383) in view of Greinke et al. (US 5,677,082), Claims 1-9, 22-25, 30-33 and 39 were rejected under 35 U.S.C. 103(a) as being unpatentable over Hase et al. in view of Tamaki et al. (US 5,698,341), Claims 1-9, 22-25, 30-33 and 39 were rejected under 35 U.S.C. 103(a) as being unpatentable over Tamaki et al. taken with Hase et al., Claims 1-9 and 30-33 were rejected under 35 U.S.C. 103(a) as being unpatentable over Hase et al. in view of Greinke et al. and further in view of Ozaki et al. (US 5,344,724), Claims 1-9, 22-25, 30-33 and 39 were rejected under 35 U.S.C. 103(a) as being unpatentable over Hase et al. in view of Tamaki et al. and further in view of Ozaki et al., Claims 1-9, 22-25, 30-33 and 39 were rejected under 35 U.S.C. 103(a) as being unpatentable over Tamaki et al. taken with Hase et al. and further in view of Ozaki et al., Claims 28-29 were rejected under 35 U.S.C. 103(a) as being unpatentable over Hase et al. in view of Greinke et al. alone, or further in view of Ozaki et al., as applied above and further in view of Yoneda et al. (US 5,591,547), Claims 28-29 were rejected under 35 U.S.C. 103(a) as being unpatentable over Hase et al. in view of Tamaki et al. alone, or further in view of Ozaki et al., as applied above and further in view of Yoneda et al., Claims 28-29 were rejected under 35 U.S.C. 103(a) as being unpatentable over Tamaki et al. taken with Hase et al. alone, or further in view of Ozaki et al., as applied above and further in view of Yoneda et al.

4. As is described in this Declaration, I conducted experiments for the purpose of demonstrating that a carbon material for a lithium battery of the present invention does not include Hase et al. (US 5,910,383).

#### A. Preparation of carbon material powder

Dehydrated coal tar was heated at 385°C for 14 hours under pressure at 3kg/cm<sup>2</sup>·G to form spherulite. The spherulite was separated from a reacted tar by a high-temperature centrifugation. 2g of the obtained spherulite was washed using 500 ml of toluene at 130°C and further washed using 40 ml of quinoline at 100°C. These washing process was conducted three times. Thereafter, the washed spherulite was dried at 150°C for 3 hours in a N<sub>2</sub> atmosphere to obtain mesophase carbon microsphere.

Amount of quinoline-insoluble components (referred to as QI) in the obtained mesophase carbon microsphere was measured by a method of JIS K2425, and the QI was 84.5%.

The obtained mesophase carbon microsphere was heated to 700°C at a rate of 0.1°C/hr in an N<sub>2</sub> atmosphere, and held for 1 hour to carbonized, thereafter, particles of mesophase carbon microsphere were grinded by a hammer mill. Obtained fine particles of mesophase carbon microsphere were subjected to a size control by an air classifier.

The fine particles of mesophase carbon microsphere were subjected to an oxidation treatment by heating from 130°C to 260°C at a rate of 4°C/hr in an air atmosphere and holding for 20 minutes.

A graphite crucible having a screw cap was filled with the mesophase carbon microsphere, and set in a horizontal tubular furnace. Air was evacuated from the furnace, and then, Ar was filled in the furnace. The mesophase carbon microsphere was heated from 25°C to 3000°C at a rate of 1000°C/hr in an Ar atmosphere, held for 10 minutes, and cooled to form a carbon material powder as a measurement sample.

#### B. Measurement of BET and Particles Size

##### B-1. Measurement of BET

The adsorption isotherm of the prepared carbon material powder was measured using a NOVA 1200 manufactured by Quantachrome, and N<sub>2</sub> gas was used as a adsorption gas. The BET specific surface area of the carbon material powder was calculated using a 3 points (multipoint) BET method.

#### B-2. Measurement of Particles Size

Particle size distribution (volume size distribution) of the carbon material powder was measured using Microtrac HRA manufactured by NIKKISO (laser diffraction particle size analyzer). From a measured particle size distribution, the particle size at 10%, 50%, and 90% of the cumulative frequency of particles (referred to as D10, D50, and D90) were calculated.

#### C. Aspect ratio

SEM image of the carbon material powder was observed, and aspect ration of the carbon material powder was calculated using the SEM image.

#### D. X-Ray Diffraction Analysis

##### D-1. Measuring apparatus (X-ray diffractometer)

Geigerflex RADB system manufactured by Rigaku

##### D-2. Method of analysis

0.64 g of the prepared carbon material powder and 0.16 g of standard silicone powder were each weighed, and mixed for 10 minutes using a planetary mill.

The mixed powder was filled in a measurement cell and the cell was set in the X-ray diffractometer.

The XRD pattern of the carbon material powder was measured under the measurement conditions as shown in Table 1.

Table 1

Measurement angle	Acceleration voltage	Acceleration current	Step	Scan speed
23-30°	40 kV	30 mA	0.01°	2.0°/min

The Co value of the carbon material powder was automatically calculated using graphitization degree data processing (Gakushin method software) for RIGAKU RINT2000.

The measurement of XRD pattern and the calculation of the Co value were carried out 5 times, and an average value of Co was determined.

Analytical data of B-D. are listed in Table 2.

Table 2

BET specific surface area (m <sup>2</sup> /g)	D10 (μm)	D50 (μm)	D90 (μm)	Aspect ratio	Co (Å)	Lc (Å)
0.73	9.5	18.5	33.6	1.75	6.747	473

The Co value of the carbon material powder was 6.747 Å, and this value was outside of the range of 6.745 Å or less of the present invention.

## E. Thermal Analysis

### E-1. Measuring apparatus

TG-DTA 6300 manufactured by Seiko Instruments (thermal analysis equipment)

### E-2. Method of analysis

Approximately 16 mg of the prepared carbon material powder was weighed in a platinum pan and set to the thermal analysis equipment. As a reference substance, aluminum oxide ( $\alpha$ -alumina, minus 75  $\mu$ m mesh) manufactured by Wako Pure Chemical Industries was used.

Heating and thermal analysis was started, while flowing air at a rate of 200 ml/min, and the carbon material powder was heated from 30°C to 900°C at a rate of 10°C/min. The measurement was carried out two times.

### E-3. Results

The measured TG/DTA profiles are shown in Fig. 1 and Fig. 2. In the Figures, line A shows DTA profile, and line B shows TG profile. TG/DTA profiles shown in Fig. 1 were measured under a condition in which weight of the carbon material powder was 16.0406 mg, and weight of  $\alpha$ -alumina as the reference substance was 12.061 mg. TG/DTA profiles shown in Fig. 2 were measured under a condition in which weight of the carbon material powder was 16.8017 mg, and weight of  $\alpha$ -alumina as the reference substance was 12.061 mg.

The oxidation initiation temperatures were 566°C and 561°C. These value were outside of the range of 600°C or more of the oxidation initiation temperature of the present invention.

Fig. 1

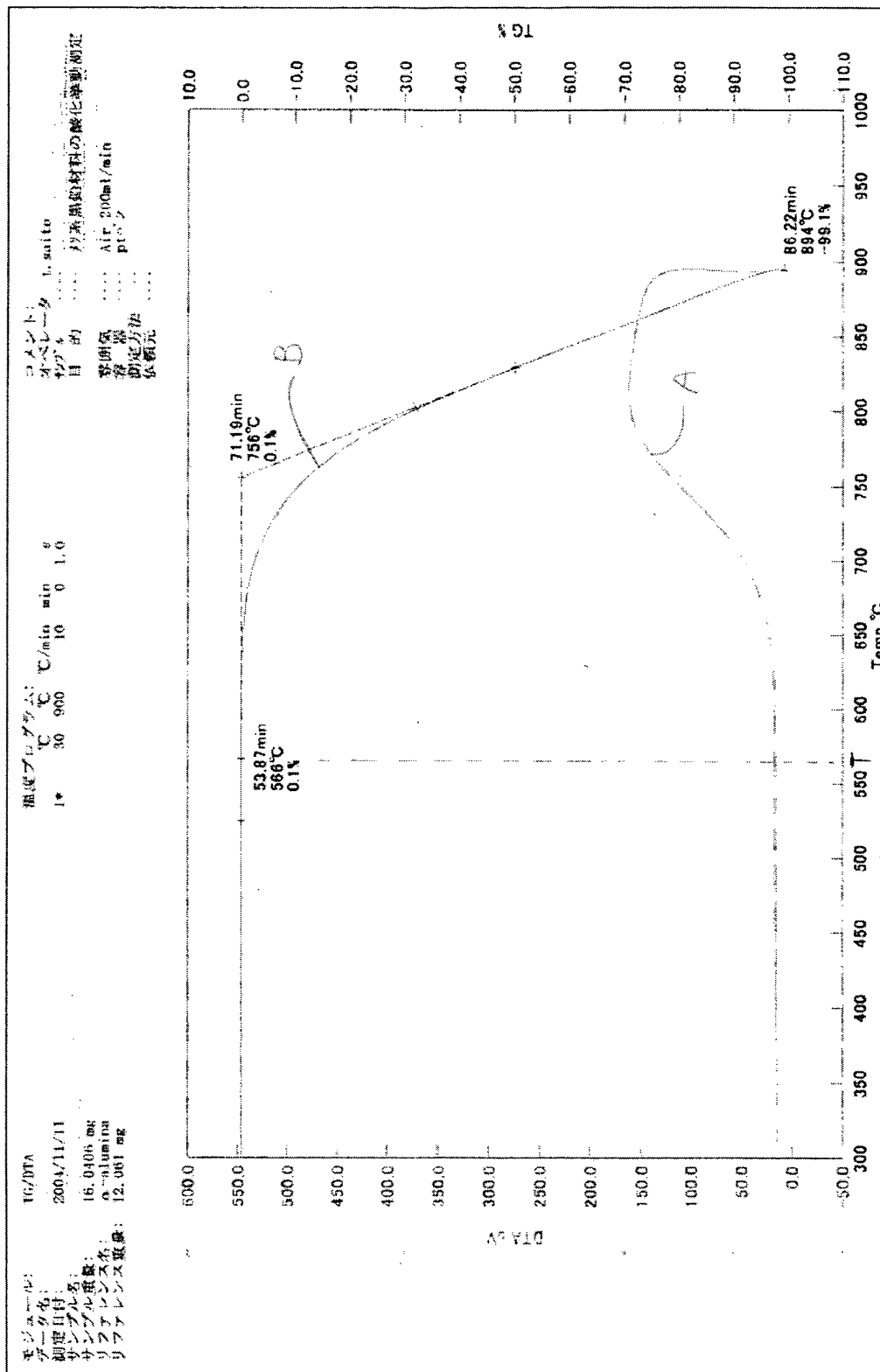


Fig.1 TG 測定





I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: 2004 / DEC / 15

Akinori Sudoh

Akinori Sudoh

BEST AVAILABLE COPY